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Sludge characteristics and performance of a membrane bioreactor for treating oily wastewater from a car wash service station

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ABSTRACT

In this study, a membrane bioreactor (MBR) with hollow fiber membrane modules was developed to potentially treat oily wastewater from a car wash station. The property and characteristics of sludge, as well as its hydrophobicity and membrane permeability, were investigated in detail. The MBR system was operated for approximately 120 d. It was found that good organic matter removal efficiencies, indicated by both COD and oil and grease results, were achieved at 90% and 88%, respectively. The specific oxygen uptake rate of the sludge in the MBR reactor was observed to be low (2.3–4.7 mg O_2/g MLVSS·h), indicating low biological activity of the sludge in the MBR system. The permeability rapidly reduced from 90 L/m^2·h·bar to below 5 L/m^2·h·bar during the operation, indicating the occurrence of membrane fouling. After acid and alkali washing, a recovery rate of over 96% of the initial membrane permeability was achieved. The results proved the ability of the MBR to treat the oily waste water from a car wash service station.

Keywords: Membrane bioreactor; Microfiltration; Fouling; Oily wastewater; Activated sludge

1. Introduction

Cars need to be washed frequently to remove dust and maintain the aesthetic appeal, while increasing their life span [1]. Washing may include the removal of coarse material, debris, dust, and oil using high pressure water and detergent mixture [2]. Washing a car requires an average of 151–227 L of water, depending on the size of the car [3]. Oil and grease are present in the car wash wastewater because vehicles generally have leaky engines. Moreover, people spray diesel or waste engine oil. Therefore, oil and grease are released from car service stations into the aquatic environment, severely damaging the surrounding ecosystems [4]. Therefore, the treatment of oily wastewater from car wash has become an urgent problem. The wastewater must be treated before being discharged. Several methods could be used for the treatment of oily wastewater, such as flotation, coagulation, biological treatment, membrane separation technology, combined technology, and advanced oxidation process. Nowadays, membrane bioreactor (MBR) is emerging as an effective technology for industrial wastewater treatment. The MBR has a high potential for the removal of petroleum and oil pollution from oil refinery wastewater treatment [5,6]. Scholz and Fuchs [7] evaluated the treatment of oily wastewater by MBR process, reporting that a high removal rate (approximately 99.99%) was achieved for fuel oil and lubricating oil. Alsalhy et al. [6] fabricated a submerged MBR with a new hollow fiber membrane for the treatment of industrial wastewater. The performance of the MBR and the removal efficiency of the oil content were investigated. It was found that the removal efficiency of the oil content was enhanced by increasing the mixed-liquor suspended solids (MLSS) concentra-

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tion. The results confirmed that the submerged MBR can offer a potential alternative process for oily wastewater treatment. The ceramic membrane with tubular configuration has also been applied in oil-in-water emulsion treatment [8]. Furthermore, Rahman and Al-Malack [9] investigated the use of a cross-flow membrane bioreactor in treating the wastewater discharged from a petroleum refinery. The performance of the MBR process was evaluated at a high MLSS concentration. The average removal efficiency of lubricating oil throughout the MBR operation was observed to be 97%. The oil refinery wastewater was also treated by submerged UF membranes. Moreover, the treatment of oil refinery wastewater using a submerged membrane bioreactor was also studied. Their results proved the potential of the MBR process in treating high-strength feed [10,11]. It should be noted that, during treatment in MBR, biodegradation is a natural process by which microbes alter and breakdown oil into other substances. The treatment efficiency depends on the micro-organisms and their capability of producing the enzymes that will degrade the target compounds. The bioavailability of weakly soluble hydrophobic compounds for microbial conversion is usually low, thus limiting their degradation rate in an aqueous medium [4]. Moreover, during the membrane filtration operation, membrane fouling is a major disadvantage, restricting the widespread application of MBR, especially in oily wastewater treatment [12]. One of the requirements in the car wash industry is that water recycling must be in accordance with the present and upcoming environmental laws. These positive evaluations imply that membrane processes can be useful in recycling wastewater in the car wash industry [13]. However, little attention has been paid to the treatment of oily wastewater from the car wash industry. The phenomena can be reflected by the limited number of articles and technical papers available in the open literature [11,13,14].

Therefore, the present study is aimed at examining the performance of oily wastewater treatment and membrane fouling control in the MBR system. In particular, the properties and characteristics of sludge, its hydrophobicity, and membrane permeability were studied.

2. Materials and methods

2.1. Cultivation of mixed bacterial culture

Three kinds of seed sludge were used for the cultivation of the mixed bacterial culture. The first sludge sample was obtained from the sludge storage tanks in the Yen So municipal wastewater treatment plant (in Hanoi, Vietnam) [15]. The second sludge sample was obtained from a pilot airlift MBR for the treatment of slaughterhouse wastewater (in Hanoi, Vietnam) [16], while the last sample was directly obtained from the mixed sludge settled on a drainage channel receiving the wastewater discharged from a car wash station (Fig. 1). The last sample could be considered as the native bacteria. The characteristics of the seed sludges are presented in Table 1.

The aim of this preliminary work is to select the most suitable mixed bacterial culture for the study. The mixed-liquor suspended solids (MLSS) concentration in the sludge seed plays an important role in cultivation. Therefore, before the cultivation experiments, the MLSS in different seed sludges (1 and 2) were maintained at the same level as seed sludge 3 (approximately 3100 mg/L) using dilution by distilled water. The wastewater from the car wash station was used for cultivation experiments. In order to enhance the cultivation process, the pH of the mineral medium ($NaNO_3$, 6.75 g/L; K_2HPO_4, 2.15 g/L; KCl, 1.13 g/L; $MgSO_4·5H_2O$, 0.54 g/L) was adjusted to 6.5 with 1.0 M HCl [17]. It should be noted that prior to use, the sludge and wastewater samples were passed through a standard sieve (0.5 mm) to remove coarse debris that could affect the cultivation process. Each seed sludge was cultivated in three 2.0-L glass cylinder beakers. The seed sludge was inoculated at 30% (volume of the seed sludge/volume of wastewater). Air was supplied by a diffuser to maintain the dissolved oxygen (DO) in the beaker within a range of 3.0–4.0 mg/L. The cultivation experiments were carried out at room temperature (25°C) for 20 d. The cultures were carried out in triplicate.

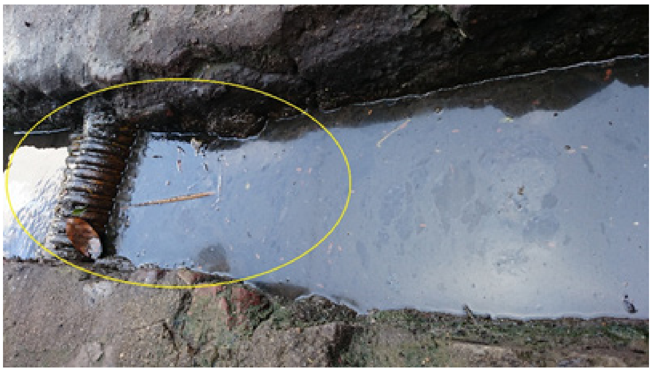

Fig. 1. Drainage system at Le Duan street (Hanoi, Vietnam).

Table 1
Characteristics of MBR membranes applied in Lab. scale reactor

Parameters	Seed sludge taken from Yen So WWTP (treating municipal wastewater) (Seed sludge 1)	Seed sludge taken from pilot-scale airlift MBR (treating slaughterhouse wastewater) (Seed sludge 2)	Seed sludge taken from a drainage (receiving wastewater from a car wash station) (Seed sludge 3)
MLSS, mg/L	4,500	8,200	3,100
MLVSS, mg/L	3,500	6,000	1,700
MLVSS/MLSS ratio (%)	0.78	0.73	0.55

2.2. Experimental MBR system

The best inoculated seed sludge was used for the study. The working volume of the MBR system used in this study (Fig. 2) was 10.5 L. A hollow fiber membrane module (PVDF), with a membrane pore size of 0.22 µm and effective membrane area of 0.065 m^2, was submerged in the bioreactor. The MBR system was operated under a constant flux (8.5 L/m^2·h) mode with 10 min of suction, followed by 2 min of relaxation. The transmembrane pressure (TMP) was continually monitored. Air was supplied continuously to the bioreactor in order to maintain the dissolved oxygen level and eliminate the effect of mixing intensity on membrane permeability. The system was operated for approximately 120 d. During the operation, if the transmembrane pressure (TMP) reached 0.66 bar (value suggested by the membrane manufacturer), the membrane module was subjected to physical cleaning (rinsing with tap water) and chemical cleaning (by immersing the membrane module for 2 h in sodium hypochloride solution, followed by 2 h in citric acid solution characterized by a pH of 2.5).

2.3. Wastewater source

The oily wastewater obtained from the drainage system receiving the wastewater discharged from a car wash station at Le Duan street (Hanoi, Vietnam) was used in this study. It contained a significant amount of large debris and sand. Therefore, it was kept in a 20-L plastic can for 2 h to let the sand settle down. Subsequently, the supernatant was filtered by a standard sieve (0.5 mm) to remove coarse debris before adding it to the reactor. The properties of this wastewater are shown in Table 2.

2.4. Hydrophobicity of sludge

The hydrophobicity of sludge (HS) was determined by a procedure based on the MATH-test [18]. Sludge samples were diluted and washed twice with a phosphate buffered saline to 2.5 g MLSS/L. This diluted sample was measured as the initial value at 650 nm in a spectrophotometer (DR2800, Hach, USA), with the filtrate from this sample being considered as a blank. Subsequently, 3 mL of this diluted sample was shaken vigorously with an equal amount of n-hexadecane for 2 min. After that, the sample was allowed to settle again for 5 min. The absorbance in the aqueous phase was subsequently measured at 650 nm (Abs_f) and compared to the absorbance of the dilution sample (as blank, Abs_i). The HS was then calculated as follows:

$$HS = 1 - (Abs_f / Abs_i) \times 100\ (\%)$$

Table 2
Characteristics of the influent wastewater used in the study

Range	pH	SS (mg/L)	COD (mg/L)	O&G** (mg/L)
Max	8.8	110	808	125
Min	8.2	51	498	37
Average	8.5	82	657	85
SD*	0.2	19	78	23

*SD: Standard deviation.
**O&G: Oil and grease.

2.5. Biodegradability of sludge

Biodegradability of the sludge samples was evaluated by the oxygen uptake rate (OUR) measurement. To compare the results from different experiments, the specific oxygen uptake rate (SOUR) was calculated. SOUR was expressed in milligrams of oxygen per gram biomass per hour as follows:

$$SOUR = \frac{OUR}{MLVSS}\ (mgO_2/gMLVSS{\cdot}h)$$

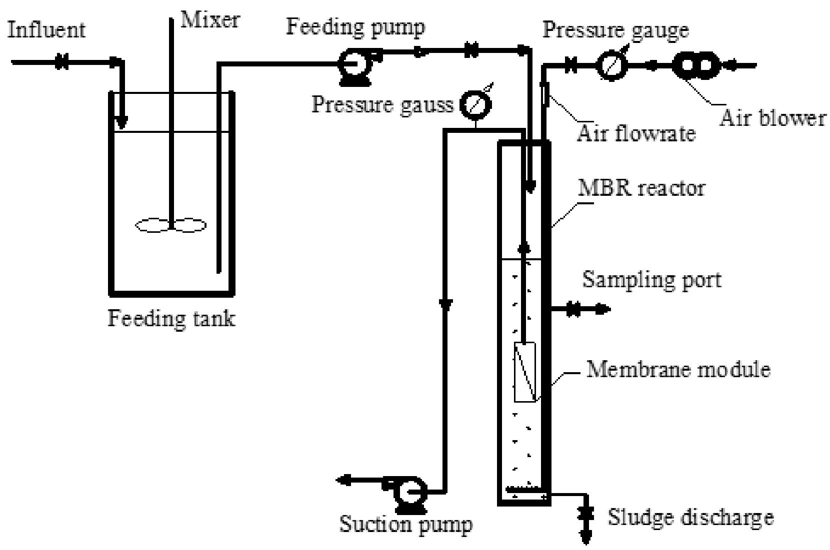

Fig. 2. Schematic diagram of the MBR system used in this study.

The mixed-liquor volatile suspended solids (MLVSS) is the volatile suspended solids concentration of the sludge sample that was used to perform the respirometric test. The procedure to measure SOUR followed the Standard Methods (method 2710 A) [19]. The DO concentration was measured by the oxygen probe.

2.6. Analytical methods

In this study, the pH was measured using a pH meter (Horiba Navi, Model F-54, Japan). DO was measured using 550A Dissolved Oxygen Instrument (YSI Inc., USA). SVI was determined by measuring the volume in mL occupied by 1 g of sludge after 30 min of settling. The transmembrane pressure (TMP) was monitored using a pressure gauge. The MLSS, MLVSS, and COD were measured based on the Standard Methods [19]. MLSS and MLVSS were measured by using the 2540D and 2540E methods, respectively. COD was determined by the closed reflux, colorimetric method (Method 8000). The total oil and grease concentration was determined by using n-hexane as the extraction solvent (USEPA Hexane Extractable Gravimetric Method 1664, adapted from Standard Methods, Section 5520B).

3. Results and discussion

3.1. Cultivation of microbial biomass

The growth profile of the three types of seed sludges are presented in Fig. 3. The profile of microbial growth during the culture time was determined through the concentration of MLSS. It should be noted that the initial MLVSS/MLSS ratios of seed sludges 1 and 2 were significantly higher than that of seed sludge 3 (0.78 and 0.73, compared with 0.55. Please see Table 1). It means the initial seed sludge 3 contains a large amount of inorganic substances. As shown in Fig. 2, during the 20 d of cultivation, the seed sludges obtained from the municipal wastewater (seed sludge 1) and a slaughterhouse wastewater treatment plant (seed sludge 2) developed very slowly. However, the seed sludge obtained from a drainage receiving wastewater from a car wash station (Seed sludge 3) showed a rapid growth during the period ranging from 6 to 10 d, after which there were no significant differences in the microbial growth used for the cultivation of the mixed bacterial culture. It was clear that the stationary phase began after 10 d of culture, at which stage, only a fraction of hydrocarbons probably remained difficult to degrade [20,21]. The results show that the native microbial biomass cultivated from the sludge obtained from a car wash wastewater drainage developed well and was able to degrade the oily wastewater. Based on the observations from the cultivation experiments, seed sludge 3 was collected and inserted into the MBR system for further study.

3.2. COD removal in the MBR system

Fig. 4 presents the COD of the influent and effluent from the MBR system. It is clear that the COD varied in a wide range. The results obtained indicated that after the first 18 days of operation, the MBR system achieved a good COD removal efficiency. A significant reduction in COD concentration was observed, with a wide range from 500–800 mg/L to lower than 70 mg/L in the MBR system, indicating a high removal efficiency of up to 90%, confirming the robustness of the MBR system for the treatment of oily wastewater. Moreover, in this study, the mixed liquid samples in the MBR reactor were sometimes filtered (0.45 µm) to obtain the supernatant to measure the soluble COD, to compare the COD removal by biological and filtration processes. The observations show that the biological contribution was approximately 86%, highlighting the significant development of biomass activity. The physical filtration membrane contributed to a further increase in the COD removal efficiency (approximately 4%) by retaining within the MBR reactor, the particles with dimensions larger than the 0.22-µm membrane pore size used in this study. In submerged MBRs, the membrane filtration plays an important role in maintaining a high and stable COD removal. It serves as an additional purification phase to retain the remaining particulate COD and biomass, thus improving the quality of the treated wastewater [11]. It should be noted that the oily wastewater was not easily biodegradable. Therefore, the biomass could not be fully acclimated to this wastewater [22,23]. As seen from Fig. 4, the typical COD values of the permeate were close to 70 mg/L most of the time, showing that the presence of the recalcitrant components were not properly disposed with biological treatment.

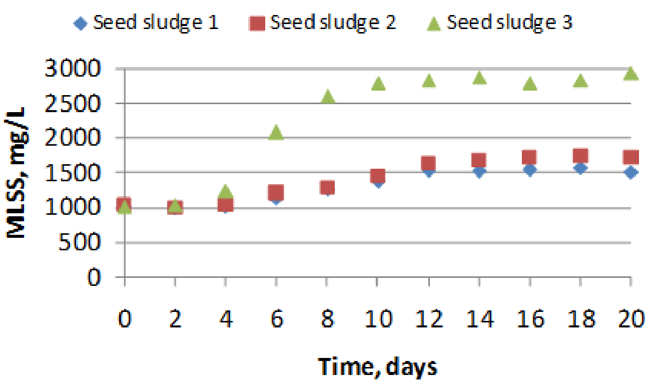

Fig. 3. Growth profile of different seed sludges during cultivation.

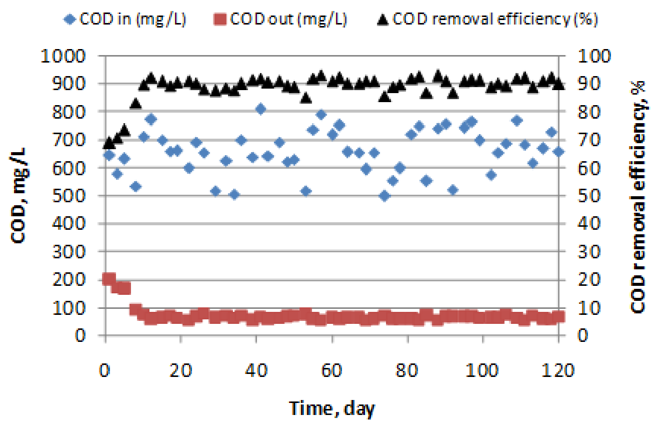

Fig. 4. Variation of the influent and effluent COD with time.

3.3. Oil and grease (O&G) removal in the MBR system

The oil and grease (O&G) concentration of the influent was approximately 35–125 mg/L (Fig. 4). As seen from Fig. 5, the O&G concentration measured in the effluent showed that it was slowly removed during first 40 d, following which its concentration declined significantly to lower than 40 mg/L. The average O&G removal rate of 88% was achieved after 50 d of operation. It should be noted that the car wash wastewater containing oil, grease, and chemical surfactants, is susceptible to microbial degradation processes [24]. Moreover, a large quantity of oil was present in both, emulsified and non-emulsified forms [4]. Therefore, biological treatment of the car wash service station wastewater is feasible with a long retention time [12]. It should be mentioned that the detergents surround oil droplets with a layer of the detergent molecule to create a water-soluble coating [14]. During treatment, the oil uptake by a microbial biomass in a bioreactor can be carried out by two mechanisms: uptake by direct interfacial contact of microorganisms with oil drops and uptake by emulsified forms of oil [21]. Scholz and Fuchs [7] studied the MBR using an ultrafiltration membrane unit, with an oil removal rate higher than 99%. A cross-flow micro filtration process was applied to treat the oily wastewater. An oil removal efficiency of higher than 92% was achieved [25]. Yang et al. [26] developed an efficient dynamic membrane for application in oily wastewater treatment to achieve oil removal of over 99%. In the case of Salahi et al. [27], a nano-porous membrane sheet was employed to treat the oily wastewater in a desalter plant. The results show that nano-porous membranes are efficient for the treatment of oily wastewater (99%) [27].

3.4. Characteristics of sludge in the MBR system

SOUR has been widely recognized as an important parameter in biomass viability monitoring [28]. In this study, the SOUR of the sludge in the MBR reactor was measured. It varied within a range of 2.3–4.7 mgO_2/gMLVSS·h (Fig. 6). The observations show that all samples presented low SOUR, well below 10 mgO_2/gMLVSS·h, indicating low biological activity of the sludge in the MBR system [28]. The low biological activity of the sludge could be attributed to the fact that the amount of 'food' available in the oily wastewater for bacteria was limited, as the measured respiration was mostly endogenous [29].

Moreover, the SOUR values were lowest during the first 10 d (Fig. 6), indicating low biological activity of the activated sludge during the initial operation. The bioactivity could be affected by low temperatures, lower hydrolysis rate, or different COD content during the operation [20,30]. In addition, low SOUR values show that a less easily biodegradable carbon source was present in the oily wastewater [31]. This is one of the factors becoming predominant in terms of biomass viability [5,22]. It should be noted that SVI is widely used for evaluating the compressibility and settleability of activated sludge. SVI provides information about the flocculation state of the activated sludge [22]. In this study, during the first 10 days, the SVI was lower than 85 mL/g, with the sludge showing high settle ability initially. After 18 d, the SVI of sludge increased to 132 mL/g (Fig. 6). Better compressibility can increase the porosity of the cake layer, thereby enhancing the membrane flux.

3.5. Membrane fouling during treatment

The variation of permeability with time is presented to assess the membrane fouling behavior in the MBR system. The permeability (P) is calculated from the following equation:

$$P = \frac{J_p}{TMP} \quad (\text{L/m}^2\cdot\text{h}\cdot\text{bar})$$

where J_p is the permeate flux (L/m²·h), and TMP is the transmembrane pressure (bar).

Membrane permeability versus time in the MBR during the operation period was determined and is presented in Fig. 7.

As shown in Fig. 7, a rapid reduction in permeability was observed, from 90 L/m²·h·bar to approximately 5 L/m²·h·bar during the first 10 d. It was induced by the adsorption of the oil-in-water emulsion with a small droplet size and pore plugging. After that, the membrane permeability was stable at approximately 4.5 L/m²·h·bar for approximately 40 d of operation. Subsequently, the fouled membranes were cleaned by physical and chemical

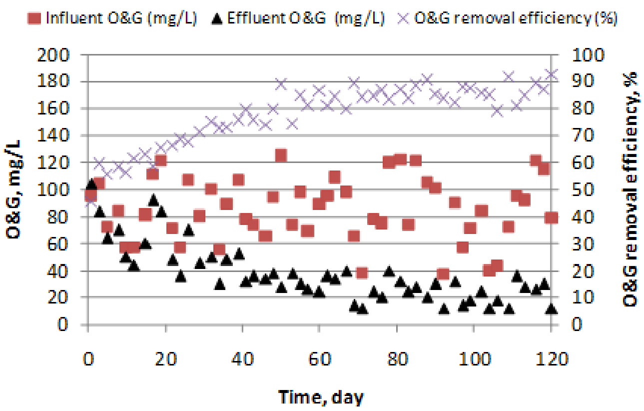

Fig. 5. Variation of the influent and effluent O&G with time.

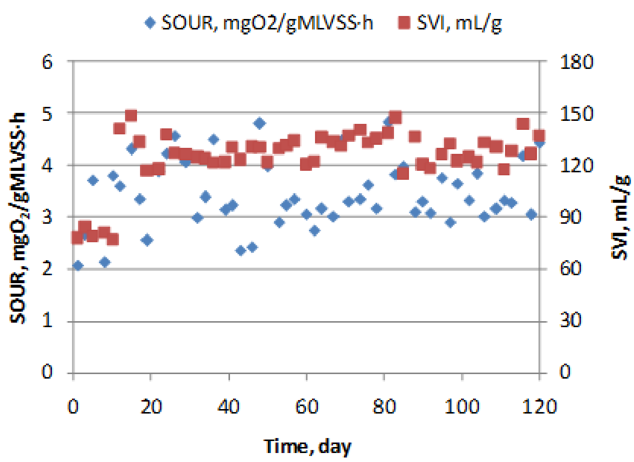

Fig. 6. Variation of SOUR and SVI during operation.

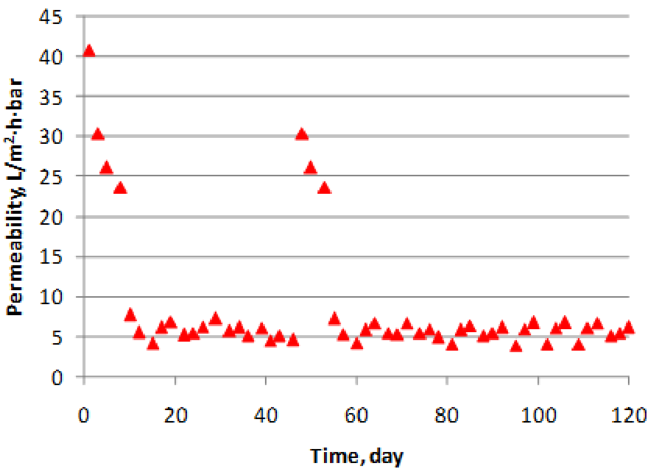

Fig. 7. Variation of permeability during operation.

methods, in that order, to remove the cake layer formation and pore blocking. After acid and alkali washing, initial membrane permeability recovery of more than 96% was obtained. In this study, the hydrophobicity of the sludge was often analyzed to investigate the correlation between its property and membrane fouling. The observed value of sludge hydrophobicity was approximately 80%. The high hydrophobicity of the sludge was caused due to the strong connection of the highly dispersed oil droplets to the sludge. It should be noted that the membranes used in MBRs are typically made hydrophilic to improve their water permeability. Low hydrophobicity of sludge flocs may cause high fouling due to stronger interactions with the membrane surface [32]. Moreover, decreasing sludge hydrophobicity results in floc deterioration, which could lead to severe membrane fouling [33].

4. Conclusions

Oily wastewater could be treated effectively by MBR technology. The average removal rates for COD and O&G were observed to be higher than 90% and 88%, respectively. Low biological activity was observed for the MBR sludge. The high hydrophobicity of the sludge was caused due to the strong connection of the highly dispersed oil droplets to the sludge, thus increasing membrane fouling. The fouled membranes could be recovered up to 96% when they were cleaned by physical and chemical methods. From the results obtained, it can be concluded that the MBR offers a potential alternative treatment process for oily wastewater from car wash service station. MBR could supply an effluent of satisfactory quality for water reuse in the car wash industry. Further study should be conducted to control the membrane fouling in order to prolong the membrane filtration period, aiming to reduce the cost of the MBR application.

Acknowledgments

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